German Joint Research Project on “Conditioning of long-lived Radionuclides in Ceramic Waste Forms”

Introduction:

The disposal of high level radioactive waste is one of the most pressing and demanding challenges of the 21st century. With respect to long-term safety aspects of geological disposal, the minor actinides (MA) such as Am, Cm and Np and long-lived fission and activation products such as ^{14}C, ^{36}Cl, ^{79}Se, ^{90}Sr, ^{99}Tc, ^{135}Cs, and ^{129}I may be of particular concern due to their long half-lives, their high radiotoxicity and mobility, respectively.

Ceramic waste forms for the immobilisation of these radionuclides have been investigated extensively in the last decades since they exhibit certain advantages compared to other waste forms (incl. borosilicate glasses and spent fuel) such as high loadings and chemical durability.

This project focuses on basic research on long-term behaviour of ceramic waste forms.

Funding:

SPONSORED BY THE

Federal Ministry of Education and Research

Support Code: 02NUK021

- Project duration: 3 years
- Funding: ~ 2.6 Mio. EUR
- 7 national partners from research centers, university and industry

Project goal:

- Fundamental studies on ceramic waste forms
- Solid state chemistry of actinides
- Correlation between structure and material science
- Networking between Research Center, university and industry
- Promoting young scientists
- Sustainability of competence

➤ Provide a refined understanding on a molecular level of ceramic materials with respect to long-term stability under conditions relevant to nuclear disposal in a deep geological repository.

Phases:

- Monazites ($LnPO_4$; Ln = La-Gd)
- Pyrochlore $(Ln,An)_2(Zr,Hf)_2O_7$
- Apatite
- Layered Double Hydroxides
- Phosphosilicates

Why ceramics?

- Chemical flexibility
- High actinide loading
- Mechanical stability
- Chemical durability

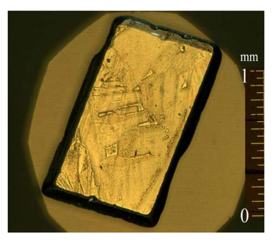
Natural Monazite: $(Ce,La,Y,Th)PO_4$

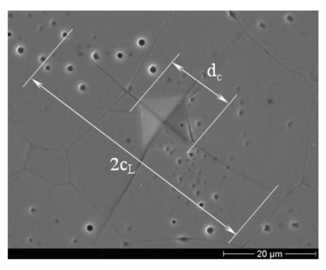
- Natural analogues contain significant amounts of Th and U without any indication of radiation damages over billions of years

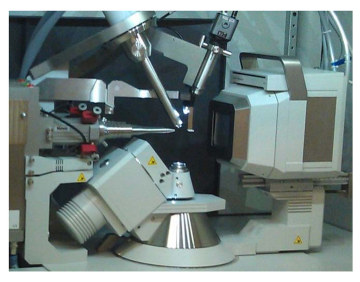
Working packages:

Physical properties

- Density, hardness, fracture toughness
- Heat capacity, elasticity
- Sinterkinetics

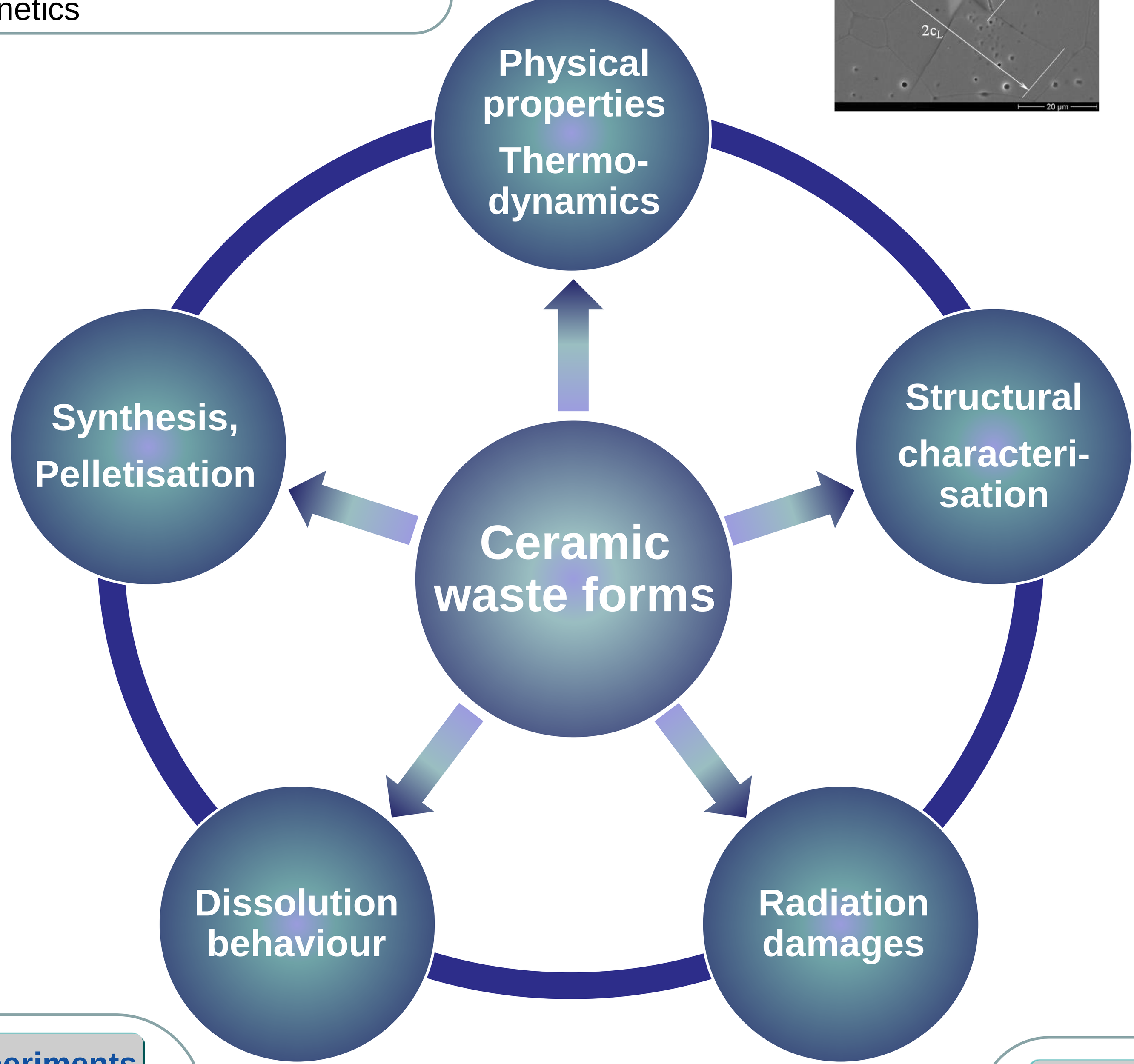






Sythesis Methods

- Precipitation
- Hydrothermal synthesis
- Sol-gel
- Flux-method (Single crystals)

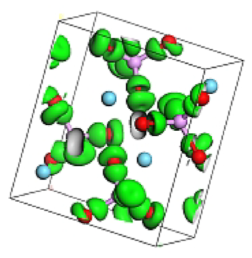


Structurlal Characterisation

- Microstructure (Optical microscope, SEM, FIB/TEM)
- Long range order (XRD)
- Short and medium range order (IR, Raman, TRLFS, EXAFS)

Molecular Modelling

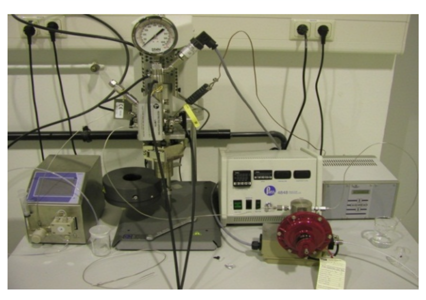
- DFT calculation → Electron density function
- Computer simulations of radiation damages (SRIM)



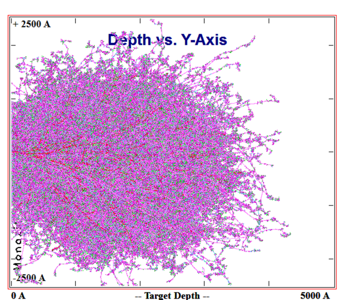
Static experiments

- Simple set-up
- Standardised tests
- Most common set-up
- Drift of pH and c (Ln^{3+})

Dynamic experiments



- Far from equilibrium
- Defined conditions
- Constant parameters



Self Irradiation

- Incorporation of short-lived actinides
- (^{238}Pu; 87.7 a half-life)
- Long-term experiment

Heavy Ion Irradiation

- Heavy ion bombardment (Xe^+, Kr^+, Ar^+, Au^+, Bi^+)
- α-irradiation; (Nuclear recoil effect neglected)
- Short-term experiment
- Surface sensitive

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